SAVANNAH RIVER NATIONAL LABORATORY
Operated by Savannah River Nuclear Solutions, LLC

We Put Science To Work

Qualification of the Savannah River Site ^{252}Cf Shuffler for Receipt Verification Measurements of Mixed U-Pu Oxides

Franklin H. DuBose, PhD
Savannah River National Laboratory

April 20, 2012

Health Physics Society

Savannah River Chapter

In this talk…

Motivation

Overview of the K-Area ^{252}Cf Shuffler

Principle of Operation

Calibration

Results

Motivation

K-Area work revolves around two programs

3013 Surveillance

Material Receipt/Storage

Both require extensive Pu measurement capabilities

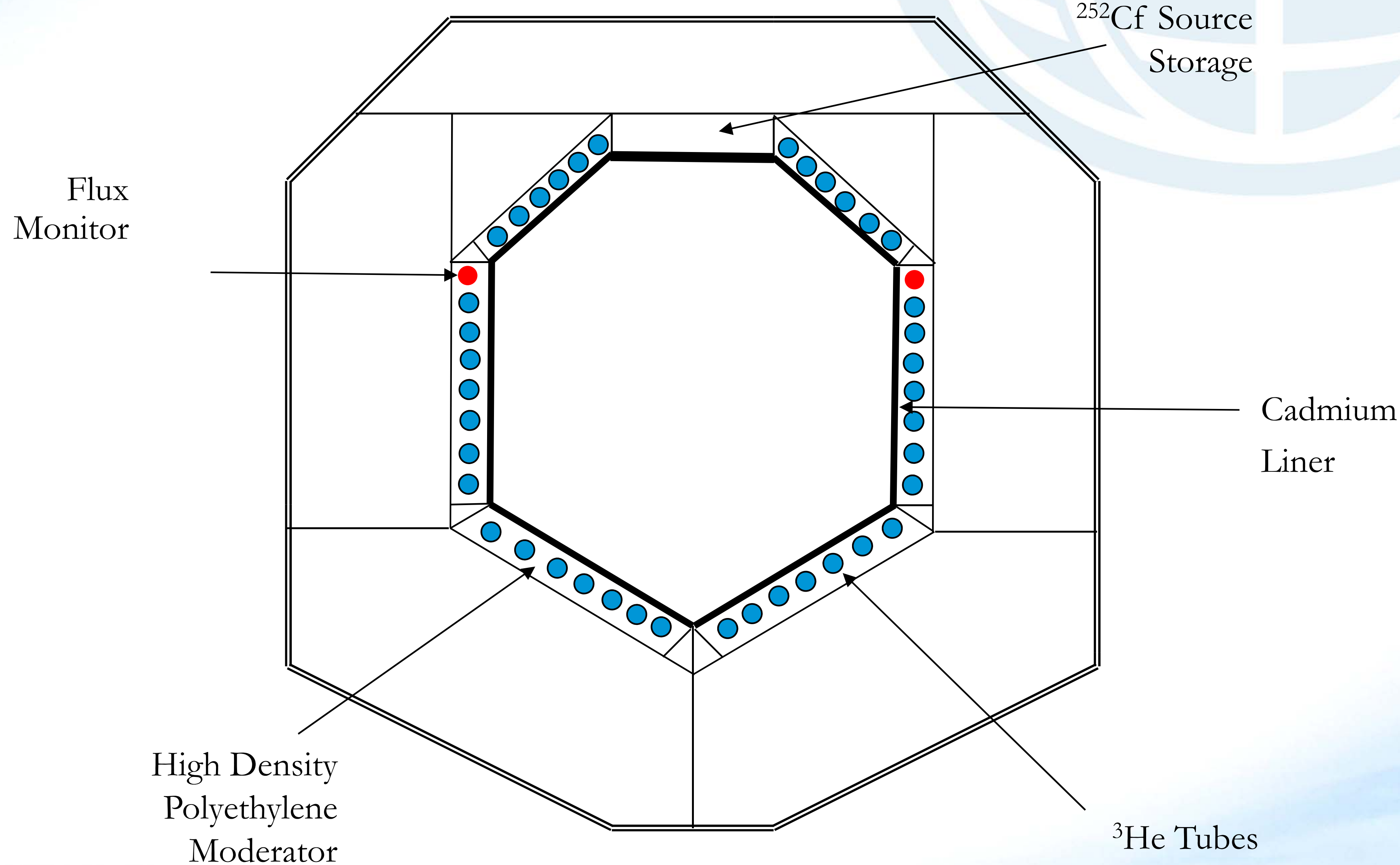
Required similar ^{235}U measurement capabilities

- Mixed Oxide
- U_3O_8
- U-metal

^{252}Cf Shuffler

- 60 10-atm ^{3}He Tubes
- 2 4-atm flux monitors
- ~600 μg ^{252}Cf source
- PLC
- JSR-15 Shift Register
- Derandomizer
- JSB-96

^{252}Cf Shuffler Layout



SRNL

Neutron Counting

Source Irradiates at ~ 10^9 n/s/g

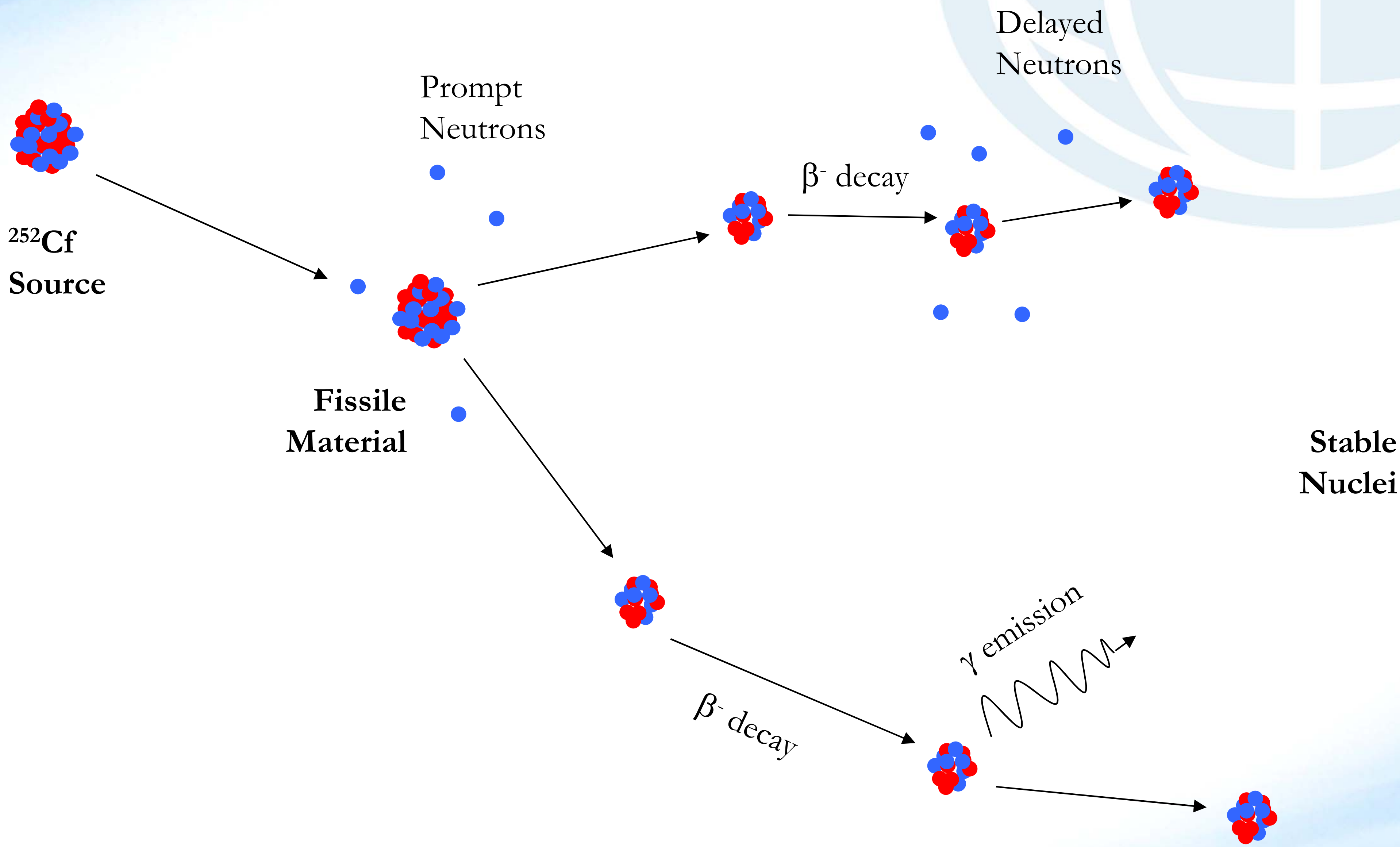
Resulting in emitted neutrons that are:

Prompt: Immediate

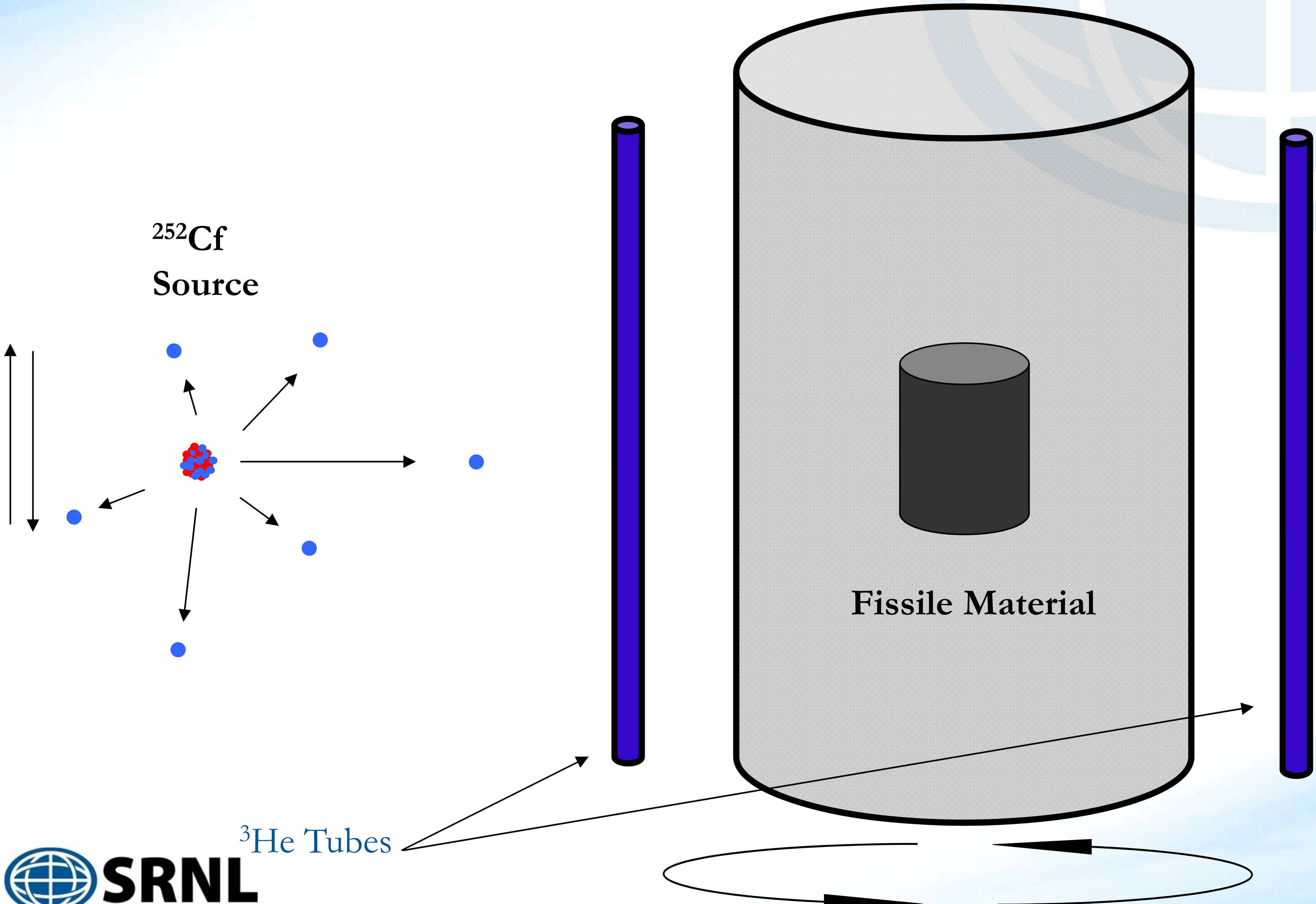
Delayed: seconds – minutes after the initial irradiating event

Delayed neutrons are counted for several seconds after removing the source.

Delayed Neutron Production



Principle of Operation



SRNL

Calibration

Determined optimal assay parameters:

- ~ 1 s Forward time/Reverse Time
- 20 s Irradiation Time
- 10 s Delayed Neutron Count Time
- 70 Shuffles

Performed simulations of selected standards: MCNPX

SRNL

Calibration Standards

High Purity

PuO_2 (5)

- 200 – 1300g
- 80 – 94% ^{239}Pu

UPu (6)

- 0.1 – 1300g Pu
- 94% ^{239}Pu
- 400 – 1700g U
- 93% enriched

Working Standards

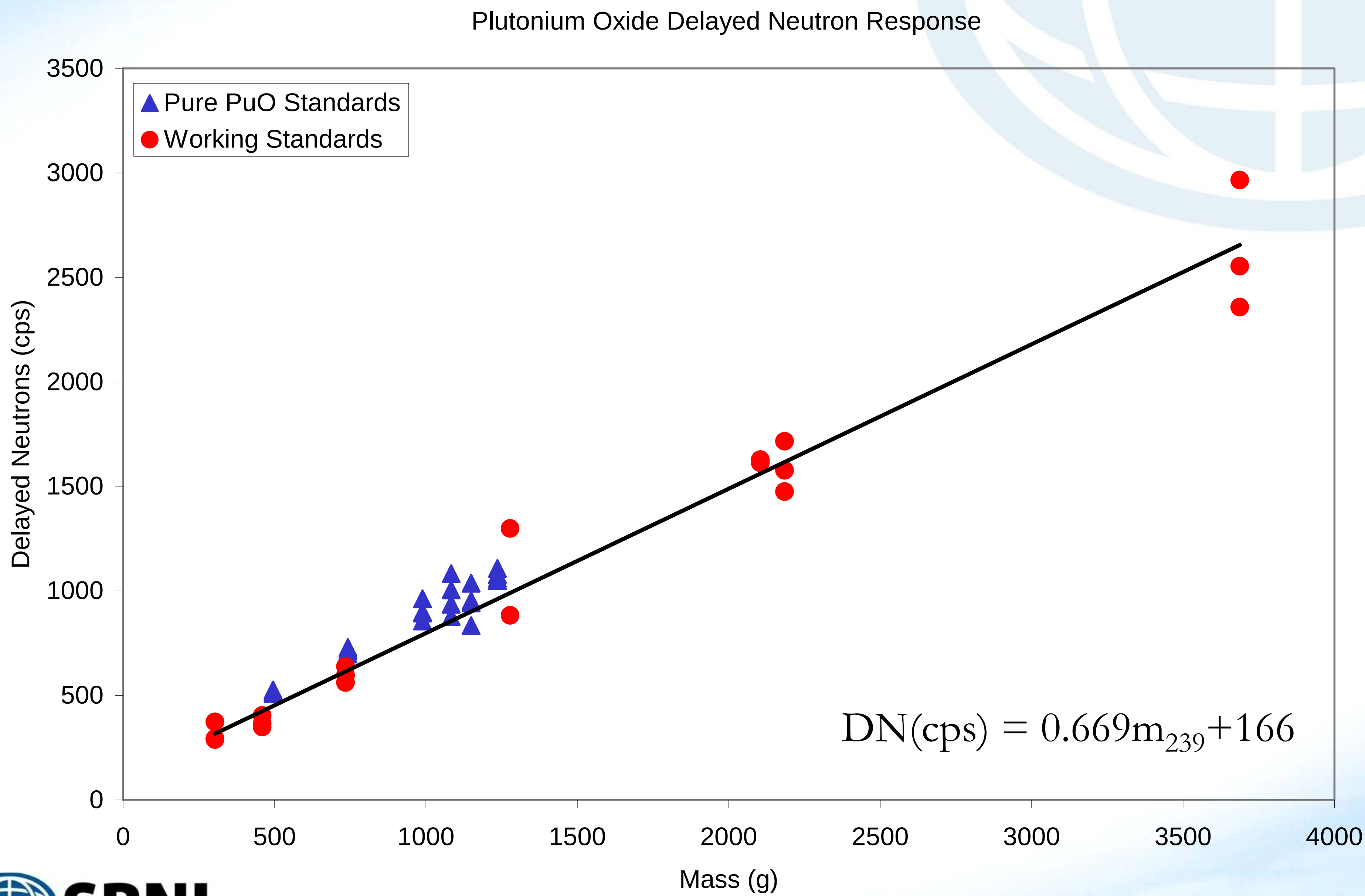
- PuO_2 (7)
  - 400 – 4000g
  - 75 – 94 % ^{239}Pu
- UPu (3)
  - 130 – 860g Pu
  - 88 – 94% ^{239}Pu
  - 1600 – 4000g U
  - 80 - 94%

SRNL

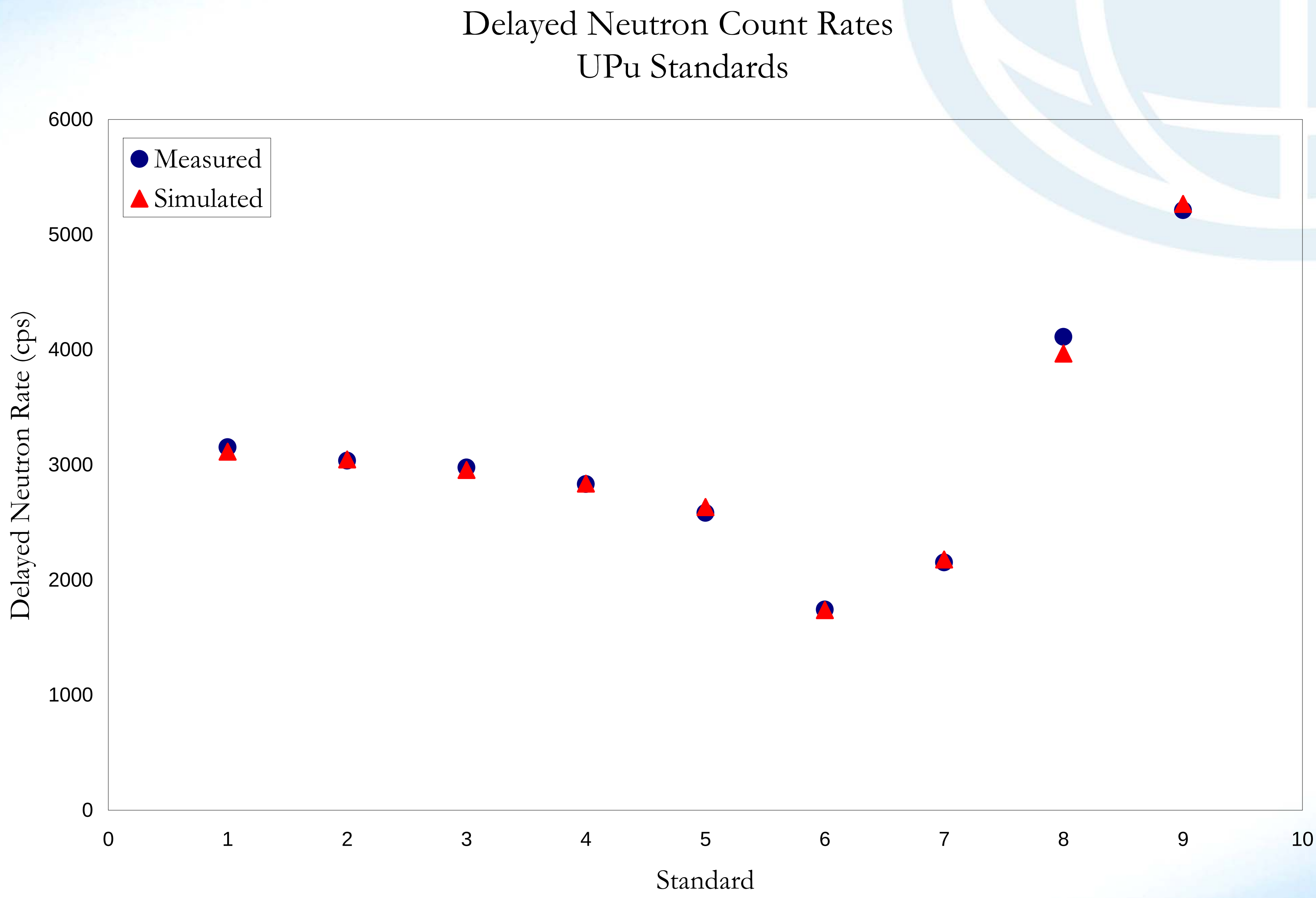
Measurement and Simulation

Measurement	Simulation
Each item measured five (3 - 5) times, with a 15 minute hold between measurements	MCNP5/MCNPX biased using time and energy dependent weight windows
Inner containers from HRS, LLNL, RFETS, LANL	Dimension of the HRS container used in all cases
Operating parameters identical for all items	Material density varied from 2.5 – 4.0 g/cc to accommodate 3013 volume

Measured PuO_2 Response



Simulated and Measured MOX Response



SRNL

Measured vs Simulated DN Rates: UPu Standards

Standard	Percent Difference
1	-1.20
2	0.30
3	-0.73
4	0.17
5	2.35
6	-0.25
7	1.18
8	-3.49
9	1.06

SRNL™

The simulations showed that…

- delayed neutron contributions ^{235}U and ^{239}Pu dominated the response
- ^{239}Pu cps/g highly dependent on relative concentration
- average delayed neutron response behavior is viable for prediction
- density must be considered

SRNL

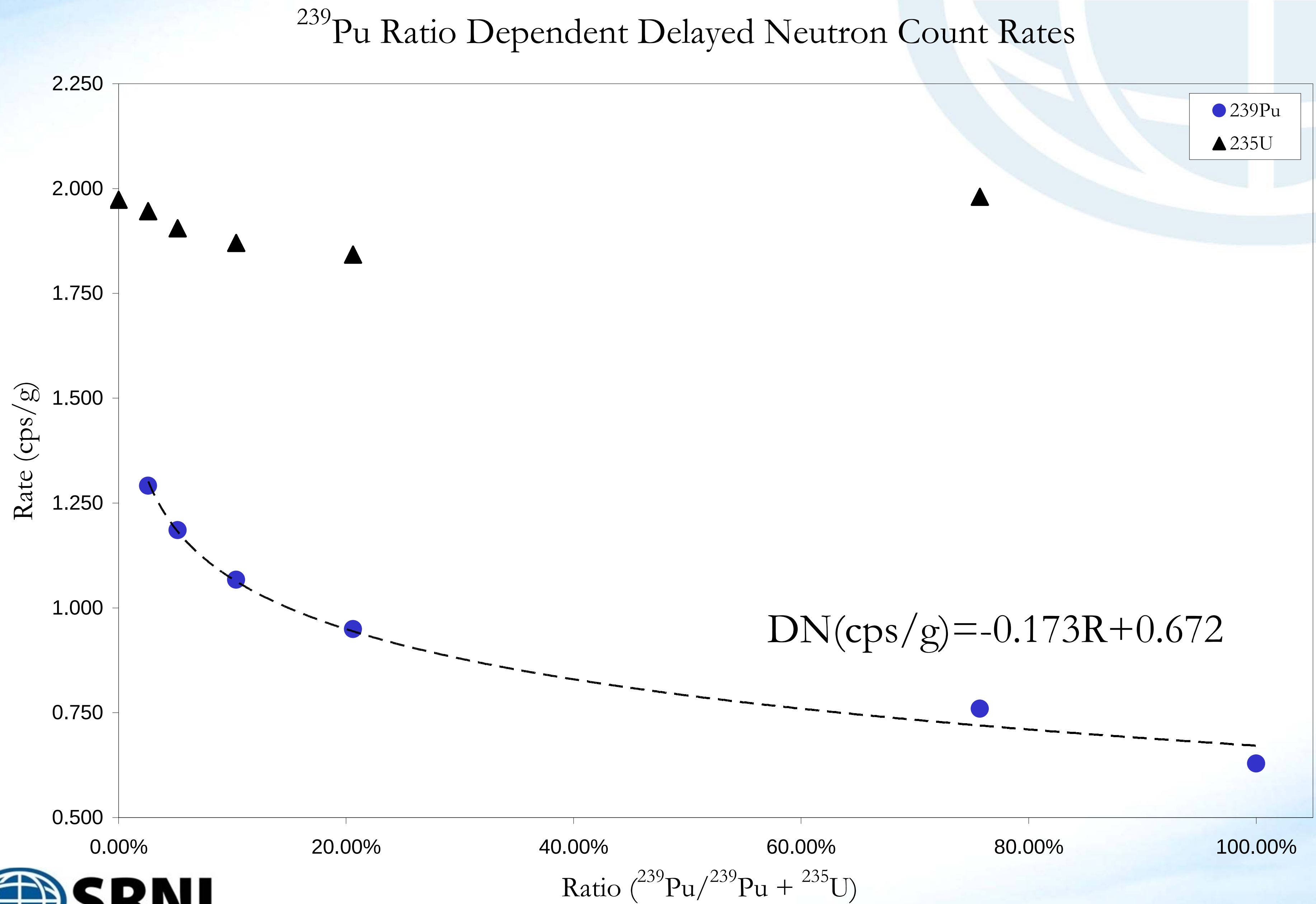
Calibration Methodologies

Adjusted Count Rate Method

- Decreases the measured delayed neutron count rate by subtracting out the ^{239}Pu activation contribution
- Determines ^{235}U content from the ^{235}U cps/g
- Limited by ^{239}Pu content

Combined Count Rate Method

- Delayed neutron count rate is a linear combination of ^{235}U and ^{239}Pu activation contributions
- Coefficients depend on material density



^{239}Pu Ratio Dependent Delayed Neutron Count Rates
2.250
2.000
1.750
1.500
1.250
1.000
0.750
0.500
Rate (cps/g)
239Pu
235U
DN(cps/g)=-0.173R+0.672
0.00%
20.00%
40.00%
60.00%
80.00%
100.00%
Ratio (^{239}Pu/^{239}Pu + ^{235}U)

SRNL

Adjusted Count Rate Method

$$DN_{adj} = DN_{Net} - \left[-0.1725 \ln(R) + 0.6716 \right] \cdot m_{239}$$

$$m_{235} = \frac{DN_{adj}}{\left[-0.0501 \ln(R) + 1.7602 \right]}$$

SRNL™

Combined Count Rate Method

$$CR_{Net} = k_{235} m_{235} + k_{239} m_{239}$$

$$m_{235} = \frac{DN_{Net} - k_{239} m_{239}}{k_{235}}$$

$$k_{235} \approx \begin{cases} 2.0\text{cps/g} & \text{, low density} \\ 1.4\text{cps/g} & \text{, high density} \end{cases}$$

SRNL

Calibration Method Comparison

Adjusted Count Rate	Combined Count Rate
High precision and accuracy over the applicable range	High precision and accuracy across the entire measured range
Under predicts ^{235}U content at higher masses	Used to successfully verify ~ 94 % of all UPu items measured to date
Less accurate with higher Pu content	Average Percent Difference: < 7.0 %
Has been successfully applied in the field on several items	

Results

Two Active calibration methods have been developed to quantify ^{235}U in mixed oxide

Method 1 is ^{239}Pu ratio dependent

Method 2 assumes independent contributions from each fissile nuclide

Both methods show high accuracy within applicable mass ranges

Method 1 and 2 have been successfully applied to verify over 100 mixed U-Pu bearing items

Planned and Completed Work

- Multiplicity characterization and measurement of total Pu in PuO_2 and Mixed Oxide – Complete
- Further assessment of ^{239}Pu ratio dependencies - Complete
- Feasibility studies for "impure" metals and high Be oxides – Complete
- Feasibility studies for known-α analysis of oxides - Complete
- Qualification for verification of total Pu in pure and impure metals – In Progress
- Qualification for verification of total Pu in impure oxides – In Progress
- Qualification for verification of total U and Pu in mixed metals – Not Started

SRNL